

## Claims

- [c1] 1.A composite article comprising a substrate having at least a substrate surface and a coating disposed on said at least a substrate surface, said coating comprising a coating material, a composition of which varies substantially continuously across a thickness of said coating.
- [c2] 2. The composite article according to claim 1, wherein said substrate comprises a polymeric material.
- [c3] 3. The composite article according to claim 2, wherein said polymeric material is selected from the group consisting of polyethyleneterephthalate, polyacrylates, polycarbonate, silicone, epoxy resins, silicone-functionalized epoxy resins, polyester, polyimide, polyetherimide, polyethersulfone, polyethylenenaphthalene, polynorbonene, poly(cyclic olefins).
- [c4] 4.The composite article according to claim 1, wherein said coating material is selected from the group consisting of organic, inorganic, ceramic materials, and combinations thereof.
- [c5] 5.The composite article according to claim 4, wherein said inorganic and ceramic materials are selected from the group consisting of oxide, nitride, carbide, boride, and combinations thereof of elements of Groups IIA, IIIA, IVA, VA, VIA, VIIA, IB, and IIB, metals of Groups IIIB, IVB, and VB, and rare-earth metals.
- [c6] 6.The composite article according to claim 1, wherein said coating material is formed by a method selected from the group consisting of plasma-enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, inductively-coupled plasma-enhanced chemical-vapor deposition, and combinations thereof.
- [c7] 7.The composite article according to claim 1, wherein said coating material is formed by expanding thermal-plasma chemical-vapor deposition.

- [c8] 8.The composite article according to claim 1, wherein said coating material is formed by radio-frequency plasma-enhanced chemical-vapor deposition.
- [c9] 9.The composite article according to claim 1, wherein a transmission rate of oxygen through said substrate coated having said coating deposited thereon is less than about  $0.1 \text{ cm}^3 / (\text{m}^2 \text{ day})$ , as measured at  $25^\circ \text{C}$  and with a gas containing 21 volume-percent oxygen.
- [c10] 10.The composite article according to claim 1, wherein a transmission rate of water vapor through said substrate coated having said coating deposited thereon is less than about  $1 \text{ g} / (\text{m}^2 \text{ day})$ , as measured at  $25^\circ \text{C}$  and with a gas having 100-percent relative humidity.
- [c11] 11.The composite article according to claim 1 further comprising a diffuse region between said substrate and said coating, said diffuse region comprising materials of both said substrate and said coating.
- [c12] 12.A composite article comprising a substrate having at least a substrate surface and a coating disposed on said at least a substrate surface; said coating comprising a coating material, a composition of which varies substantially continuously across a thickness of said coating; said substrate comprising a polymeric material; said coating material comprising a material selected from organic, inorganic, ceramic materials, and combinations thereof; wherein a transmission rate of oxygen through said substrate coated having said coating deposited thereon is less than about  $0.1 \text{ cm}^3 / (\text{m}^2 \text{ day})$ , as measured at  $25^\circ \text{C}$  and with a gas containing 21 volume-percent oxygen, a transmission rate of water vapor through said substrate coated with said coating is less than about  $1 \text{ g} / (\text{m}^2 \text{ day})$ , as measured at  $25^\circ \text{C}$  and with a gas having 100-percent relative humidity, and said coating is deposited on said substrate by a method selected from the group consisting of plasma-enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, inductively-coupled plasma-enhanced chemical-vapor deposition, and combinations thereof.

- [c13] 13.A method for making a composite article, said method comprising:  
providing a substrate having at least a substrate surface;  
depositing a coating material having a composition on said substrate surface;  
and  
changing said composition of said coating material substantially continuously  
while said coating is being formed such that said composition varies  
substantially continuously across a thickness of said coating.
- [c14] 14.The method according to claim 13, wherein said depositing is selected from  
the group consisting of plasma-enhanced chemical-vapor deposition, radio-  
frequency plasma-enhanced chemical-vapor deposition, expanding thermal-  
plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-  
cyclotron-resonance plasma-enhanced chemical-vapor deposition, inductively-  
coupled plasma-enhanced chemical-vapor deposition, and combinations  
thereof.
- [c15] 15.The method according to claim 13, wherein said substrate comprises a  
polymeric material selected from the group consisting of  
polyethyleneterephthalate, polyacrylates, polycarbonate, silicone, epoxy resins,  
silicone-functionalized epoxy resins, polyester, polyimide, polyetherimide,  
polyethersulfone, polyethylenenaphthalene, polynorbone, and poly(cyclic  
olefins).
- [c16] 16.The method according to claim 13, wherein said coating material is selected  
from the group consisting of organic, inorganic, and ceramic materials.
- [c17] 17.The composite article according to claim 16, wherein said inorganic and  
ceramic materials are selected from the group consisting of oxide, nitride,  
carbide, boride, and combinations thereof of elements of Groups IIA, IIIA, IVA,  
VA, VIA, VIIA, IB, and IIB, metals of Groups IIIB, IVB, and VB, and rare-earth  
metals.
- [c18] 18.The method according to claim 13 further comprising effecting a penetration  
of at least a portion of said coating material into said substrate to produce a  
diffuse region between said substrate and said coating.

- [c19] 19.The method according to claim 18, wherein said diffuse region is produced by an energetic ion bombardment of a surface of said substrate to sputter a portion of a material of said substrate, and depositing a mixed material comprising sputtered substrate material and another material.
- [c20] 20.The method according to claim 13, wherein said changing said composition of said coating is carried out by changing a composition of reactive species generated by a plasma that is directed at said substrate.
- [c21] 21.A light-emitting device comprising:  
a flexible substantially transparent substrate having a first substrate surface and a second substrate surface, at least one of said substrate surface being coated with a graded-composition barrier coating, a composition of which varies substantially continuously across a thickness thereof; and  
an organic electroluminescent ("EL") member which comprises an organic EL layer disposed between two electrodes and is disposed on said flexible substantially transparent substrate.
- [c22] 22.The light-emitting device according to claim 21 further comprising a substantially transparent film having a second graded-composition barrier coating disposed thereon, said substantially transparent film being disposed on said organic EL member opposite to said flexible transparent substrate.
- [c23] 23.The light-emitting device according to claim 21, wherein said flexible substantially transparent substrate comprises a polymeric material selected from the group consisting of polyethyleneterephthalate, polyacrylates, polycarbonate, silicone, epoxy resins, silicone-functionalized epoxy resins, polyester, polyimide, polyetherimide, polyethersulfone, polyethylenenaphthalene, polynorbonene, and poly(cyclic olefins).
- [c24] 24.The light-emitting device according to claim 21, wherein said coating material is selected from the group consisting of organic, inorganic, ceramic materials, and combinations thereof.
- [c25] 25.The light-emitting device according to claim 24, wherein said inorganic and ceramic materials are selected from the group consisting of oxide, nitride,

carbide, boride, and combinations thereof of elements of Groups IIA, IIIA, IVA, VA, VIA, VIIA, IB, and IIB, metals of Groups IIIB, IVB, and VB, and rare-earth metals.

- [c26] 26.The light-emitting device according to claim 21 further comprising a reflective layer disposed on said organic EL layer, said reflective layer comprising a material selected from the group consisting of metals, metal oxides, metal nitrides, metal carbides, metal oxynitrides, metal oxycarbides, and combinations thereof.
- [c27] 27.The light-emitting device according to claim 21, wherein said organic EL layer comprises a material selected from the group consisting of poly(n-vinylcarbazole), poly(alkylfluorene), poly(paraphenylene), polysilanes, derivatives thereof, mixtures thereof, and copolymers thereof.
- [c28] 28.The light-emitting device according to claim 21, wherein said organic EL layer comprises a material selected from the group consisting of 1,2,3-tris{n-(4-diphenylaminophenyl) phenylamino} benzene, phenylanthracene, tetraarylethene, coumarin, rubrene, tetraphenylbutadiene, anthracene, perylene, coronene, aluminum-(picolymethylketone)-bis{2,6-di(t-butyl)phenoxides}, scandium-(4-methoxy-picolymethylketone)-bis(acetylacetonate), aluminum-acetylacetonate, gallium-acetylacetonate, and indium-acetylacetonate.
- [c29] 29.The light-emitting device according to claim 21 further comprising a light-scattering layer, said layer comprising scattering particles dispersed in a substantially transparent matrix and being disposed on a surface of said substrate opposite to said organic EL member.
- [c30] 30.The light-emitting device according to claim 29 further comprising particles of a photoluminescent ("PL") material mixed with scattering particles in said light-scattering layer, wherein said PL material is selected from the group consisting of  $(Y_{1-x}Ce_x)_3Al_5O_{12}$ ;  $(Y_{1-x-y}Gd_xCe_y)_3Al_5O_{12}$ ;  $(Y_{1-x}Ce_x)_3(Al_{1-y}Ga_y)O_{12}$ ;  $(Y_{1-x-y}Gd_xCe_y)(Al_{5-z}Ga_z)O_{12}$ ;  $(Gd_{1-x}Ce_x)Sc_2Al_3O_{12}$ ;  $Ca_8Mg(SiO_4)_4Cl_2$ ;  $Eu^{2+}, Mn^{2+}$ ;  $GdBO_3$ ;  $Ce^{3+}$ ,  $Tb^{3+}$ ;  $CeMgAl_{11}O_{16}$ ;  $Tb^{3+}$ ;  $Y_2SiO_5$ ;  $Ce^{3+}$ ,  $Tb^{3+}$ ;  $BaMg_2Al_{16}O_{40}$

$27 \text{ :Eu}^{2+}, \text{Mn}^{2+}; \text{Y}_2\text{O}_3 \text{ :Bi}^{3+}, \text{Eu}^{3+}; \text{Sr}_2\text{P}_2\text{O}_7 \text{ :Eu}^{2+}, \text{Mn}^{2+}; \text{SrMgP}_2\text{O}_7 \text{ :Eu}^{2+}, \text{Mn}^{2+}; (\text{Y,Gd})(\text{V,B})\text{O}_4 \text{ :Eu}^{3+}; 3.5\text{MgO}.0.5\text{MgF}_2 \cdot \text{GeO}_2 \text{ :Mn}^{4+}$   
 (magnesium fluorogermanate);  $\text{BaMg}_2\text{Al}_{16}\text{O}_{27} \text{ :Eu}^{2+}; \text{Sr}_5(\text{PO}_4)_4\text{Cl}$   
 $2 \text{ :Eu}^{2+}; (\text{Ca,Ba,Sr})(\text{Al,Ga})_2\text{S}_4 \text{ :Eu}^{2+}; (\text{Ba,Ca,Sr})_5(\text{PO}_4)_4(\text{Cl,F})_2 \text{ :Eu}^{2+}, \text{Mn}^{2+}; \text{Lu}_3\text{Al}_5\text{O}_{12} \text{ :Ce}^{3+}; \text{Tb}_3\text{Al}_5\text{O}_{12} \text{ :Ce}^{3+}$ ; and mixtures thereof; wherein  $0 \leq x \leq 1$ ,  $0 \leq y \leq 1$ ,  $0 \leq z \leq 5$  and  $x+y \leq 1$ .

- [c31] 31. The light-emitting device according to claim 29 further comprising at least an organic PL material dispersed in said scattering layer, said organic PL material being capable of absorbing at least a portion of electromagnetic ("EM") radiation emitted by said organic EL material and emitting EM radiation in a visible spectrum.
- [c32] 32. The light-emitting device according to claim 21, wherein said organic EL member further comprises at least an additional layer disposed between one of said electrodes and said organic EL layer, said additional layer performing at least a function selected from the group consisting of electron injection enhancement, electron transport enhancement, hole injection enhancement, and hole transport enhancement.
- [c33] 33. A light-emitting device comprising:  
 a flexible substantially transparent substrate having a first substrate surface and a second substrate surface, at least one of said substrate surface being coated with a graded-composition barrier coating, a composition of which varies substantially continuously across a thickness thereof; and  
 an organic electroluminescent ("EL") member which comprises an organic EL layer disposed between two electrodes and is disposed on said flexible substantially transparent substrate;  
 wherein said flexible substantially transparent substrate comprises a polymeric material selected from the group consisting of polyethyleneterephthalate, polyacrylates, polycarbonate, silicone, epoxy resins, silicone-functionalized epoxy resins, polyester, polyimide, polyetherimide, polyethersulfone, polyethylenenaphthalene, polynorbonene, and poly(cyclic olefins); said coating material is selected from the group consisting of organic, inorganic, ceramic

materials, and combinations thereof; and said organic EL layer comprises a material selected from the group consisting of poly(n-vinylcarbazole), poly(alkylfluorene), poly(paraphenylene), polysilanes, derivatives thereof, mixtures thereof, copolymers thereof, 1,2,3-tris{n-(4-diphenylaminophenyl)phenylamino} benzene, phenylanthracene, tetraarylethene, coumarin, rubrene, tetraphenylbutadiene, anthracene, perylene, coronene, aluminum-(picolymethylketone)-bis{2,6-di(t-butyl)phenoxides}, scandium-(4-methoxy-picolymethylketone)-bis(acetylacetonate), aluminum-acetylacetonate, gallium-acetylacetonate, and indium-acetylacetonate.

[c34]

34.A light-emitting device comprising:

a flexible substantially transparent substrate having a first substrate surface and a second substrate surface, at least one of said substrate surface being coated with a first graded-composition barrier coating, a composition of which varies substantially continuously across a thickness thereof;

an organic electroluminescent ("EL") member which comprises an organic EL layer disposed between two electrodes and is disposed on said flexible substantially transparent substrate;

a reflective layer disposed on said organic EL member opposite to said substrate; and

a substantially transparent film having second graded-composition barrier coating disposed on said reflective layer opposite to said organic EL member; wherein said flexible substantially transparent substrate and said substantially transparent film comprise a polymeric material selected from the group consisting of polyethyleneterephthalate, polyacrylates, polycarbonate, silicone, epoxy resins, silicone-functionalized epoxy resins, polyester, polyimide, polyetherimide, polyethersulfone, polyethylenenaphthalene, polynorbonene, and poly(cyclic olefins); said first and second graded-composition barrier coating material comprise a material independently selected from the group consisting of organic, inorganic, ceramic materials, and combinations thereof; and said organic EL layer comprises a material selected from the group consisting of poly(n-vinylcarbazole), poly(alkylfluorene), poly(paraphenylene), polysilanes, derivatives thereof, mixtures thereof, copolymers thereof, 1,2,3-tris{n-(4-

diphenylaminophenyl) phenylamino} benzene, phenylanthracene, tetraarylethene, coumarin, rubrene, tetraphenylbutadiene, anthracene, perylene, coronene, aluminum-(picolymethylketone)-bis{2,6-di(t-butyl)phenoxides}, scandium-(4-methoxy-picolymethylketone)-bis(acetylacetonate), aluminum-acetylacetonate, gallium-acetylacetonate, and indium-acetylacetonate.

- [c35] 35.The light-emitting device according to claim 34 further comprising a scattering layer disposed on said substantially transparent substrate opposite to said organic EL member, said scattering layer comprising scattering particles and particles of a PL material dispersed in a substantially transparent matrix.
- [c36] 36.A device assembly comprising a device disposed on a flexible substantially transparent substrate, said substrate having a first substrate surface and a second substrate surface, at least one of said substrate surfaces being coated with a graded-composition barrier coating, a composition of which varies substantially continuous across a thickness thereof.
- [c37] 37.The device assembly according to claim 36, wherein said device is selected from the group consisting of liquid crystal displays, a photovoltaic cells, integrated circuits, and components of medical diagnostic systems.
- [c38] 38. A device assembly comprising a device, at least a surface of which is coated with a graded-composition barrier coating, a composition of which varies across a thickness thereof.
- [c39] 39.The device assembly according to claim 38, wherein said device is selected from the group consisting of liquid crystal displays, a photovoltaic cells, integrated circuits, and components of medical diagnostic systems.
- [c40] 40.A method for making a light-emitting device, said method comprising: providing a flexible substantially transparent substrate having a first substrate surface and a second substrate surface, at least one of said substrate surface being coated with a first graded-composition barrier coating, a composition of which varies substantially continuously across a thickness thereof; and disposing an organic EL member which comprises an organic EL layer disposed between two electrodes on said flexible substantially flexible substrate.



- [c41] 41.The method for making a light-emitting device according to claim 40, wherein,said disposing said organic EL member comprises forming a first electrode by depositing a first electrically conducting material on said graded-composition barrier coating; depositing said organic EL layer on said first electrode; and forming a second electrode by depositing a second electrically conducting material on said organic EL layer.
- [c42] 42.The method for making a light-emitting device according to claim 40 further comprising disposing a reflective layer on said organic EL member opposite to said substantially transparent substrate.
- [c43] 43.The method for making a light-emitting device according to claim 42 further comprising disposing a substantially transparent film that is coated with a second graded-composition barrier coating on said reflective layer.
- [c44] 44.The method for making a light-emitting device according to claim 40 further comprising disposing a scattering layer on a surface of said substrate, said EM-radiation.conversion layer comprising particles of a PL material dispersed in a substantially transparent matrix.
- [c45] 45.The method for making a light-emitting device according to claim 40, further comprising disposing a second graded-barrier coating on said organic EL member opposite to said substantially transparent substrate.
- [c46] 46.The method for making a light-emitting device according to claim 40, further comprising disposing a second flexible substrate on said organic EL member, said second substrate having a second graded-composition barrier coating thereon.
- [c47] 47.A method for making a light-emitting device, said method comprising:  
providing a flexible substantially transparent substrate having a first substrate surface and a second substrate surface;  
depositing a first graded-composition barrier coating on at least one of said substrate surface, a composition of said first barrier coating varying substantially continuously across a thickness thereof, said depositing being carried out by a method selected from the group consisting of plasma-

enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, and inductively-coupled plasma-enhanced chemical-vapor deposition;

disposing an organic EL member which comprises an organic EL layer disposed between two electrodes on said flexible substantially flexible substrate; and disposing a substantially transparent film that is coated with a second graded-composition barrier coating on said organic EL member, said second graded-composition barrier coating having a composition that varies substantially continuously across a thickness thereof and being deposited on said film by a method selected from the group consisting of plasma-enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, inductively-coupled plasma-enhanced chemical-vapor deposition, and combinations thereof.

[c48] 48. The method for making a light-emitting device according to claim 47 further comprising disposing a reflective layer between said organic EL member and said coated substantially transparent film.

[c49] 49. The method for making a light-emitting device according to claim 47, wherein said depositing a first graded-composition barrier coating on at least one of said substrate surface is carried out such that at least a portion of a material of said coating penetrates into said substrate.

[c50] 50. A method for making an assembly comprising a device, said method comprising:  
 providing a flexible substantially transparent substrate having a first substrate surface and a second substrate surface, at least one of said substrate surface being coated with a first graded-composition barrier coating, a composition of which varies substantially continuously across a thickness thereof; and disposing said device on said flexible substantially flexible substrate.

- [c51] 51.The method according to claim 50, wherein said device is selected from the group consisting of liquid crystal displays, photovoltaic cells, integrated circuits, and components of medical diagnostic systems.
- [c52] 52.A method for making a device, said method comprising:  
providing a flexible substantially transparent substrate having a first substrate surface and a second substrate surface;  
depositing a first graded-composition barrier coating on at least one of said substrate surface, a composition of said first barrier coating varying substantially continuously across a thickness thereof, said depositing being carried out by a method selected from the group consisting of plasma-enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, and inductively-coupled plasma-enhanced chemical-vapor deposition;  
disposing device on said flexible substantially flexible substrate; and  
disposing a substantially transparent film that is coated with a second graded-composition barrier coating on said organic EL member, said second graded-composition barrier coating having a composition that varies substantially continuously across a thickness thereof and being deposited on said film by a method selected from the group consisting of plasma-enhanced chemical-vapor deposition, radio-frequency plasma-enhanced chemical-vapor deposition, expanding thermal-plasma chemical-vapor deposition, sputtering, reactive sputtering, electron-cyclotron-resonance plasma-enhanced chemical-vapor deposition, inductively-coupled plasma-enhanced chemical-vapor deposition, and combinations thereof.
- [c53] 53.The method according to claim 52, wherein said device is selected from the group consisting of liquid crystal displays, photovoltaic cells, integrated circuits, and components of medical diagnostic systems.